

## Accepted Manuscript

Gasification of torrefied *Miscanthus* × *gigantenus* in an air-blown bubbling fluidized bed gasifier

G. Xue, M. Kwapinska, A. Horvat, W. Kwapinski, L.P.L.M. Rabou, S. Dooley, K.M. Czajka, J.J. Leahy

PII: S0960-8524(14)00265-X

DOI: <http://dx.doi.org/10.1016/j.biortech.2014.02.094>

Reference: BITE 13092

To appear in: *Bioresource Technology*

Received Date: 6 December 2013

Revised Date: 19 February 2014

Accepted Date: 21 February 2014



Please cite this article as: Xue, G., Kwapinska, M., Horvat, A., Kwapinski, W., Rabou, L.P.L.M., Dooley, S., Czajka, K.M., Leahy, J.J., Gasification of torrefied *Miscanthus* × *gigantenus* in an air-blown bubbling fluidized bed gasifier, *Bioresource Technology* (2014), doi: <http://dx.doi.org/10.1016/j.biortech.2014.02.094>

This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

# Gasification of torrefied *Miscanthus×giganteus* in an air-blown bubbling fluidized bed gasifier

G. Xue<sup>1</sup>, M. Kwapinska<sup>2</sup>, A. Horvat<sup>1</sup>, W. Kwapinski<sup>1</sup>, L.P.L.M. Rabou<sup>3</sup>, S.

Dooley<sup>1</sup>, K. M. Czajka<sup>4</sup>, J. J Leahy<sup>\*1</sup>

1. Carbolea, University of Limerick, Ireland

2. Competence Centre for Biorefining & Biofuels, University of Limerick, Ireland

3. Energy Research Centre of Netherlands (ECN), Biomass & Energy Efficiency, Petten, Netherlands

4. Energy Engineering and Technology Division, Wroclaw University of Technology, Poland

\* Corresponding author. E-mail: [j.j.leahy@ul.ie](mailto:j.j.leahy@ul.ie); tel. : +353 61 20 2649 ; fax : +353 61 20 2568

## Abstract:

Torrefaction is suggested to be an effective method to improve the fuel properties of biomass and gasification of torrefied biomass should provide a higher quality product gas than that from unprocessed biomass. In this study, both raw and torrefied *Miscanthus×giganteus* (M×G) were gasified in an air-blown bubbling fluidized bed (BFB) gasifier using olivine as the bed material. The effects of equivalence ratio (ER) (0.18-0.32) and bed temperature (660-850 °C) on the gasification performance were investigated. The results obtained suggest the optimum gasification conditions for the torrefied M×G are ER 0.21 and 800 °C. The product gas from these process conditions

had a higher heating value (HHV) of  $6.70 \text{ MJ/m}^3$ , gas yield  $2 \text{ m}^3/\text{kg}_{\text{biomass}}$  ( $\text{H}_2$  8.6%,  $\text{CO}$  16.4% and  $\text{CH}_4$  4.4% ) and cold gas efficiency 62.7%. The comparison between raw and torrefied M×G indicates that the torrefied M×G is more suitable BFB gasification.

**Keywords:** biomass, miscanthus, torrefaction, bubbling fluidized bed gasification, mass balance

## 1. Introduction

Torrefaction is being widely investigated as a promising thermal pre-treatment method to upgrade the fuel properties of biomass prior to further thermo chemical conversion. The majority of published reports on torrefaction investigate the influence of torrefaction conditions on the fuel properties of biomass, for instance the moisture content of biomass decreases after torrefaction which can increase the energy efficiency of gasification processes as extra energy is needed to evaporate the moisture and maintain the appropriate temperature in the gasifier (Van der Stelt et al., 2011). Unstable volatile compounds, also known as tar precursors (Sweeney, 2012), can be decomposed during torrefaction which implies torrefaction could potentially decrease the tar content in the product gas from a gasification process. The energy consumption required for biomass grinding is also significantly reduced after the biomass is torrefied (Phanphanich & Mani, 2011) which is important for those applications where a reduction of the fuel particle size is needed. The energy density of torrefied biomass is enhanced due to the reduced O/C and H/C in the biomass (Bridgeman et al., 2008). Additionally, it is reported that the surface area and porosity of biomass changes with the conditions of torrefaction (Chen et al., 2011a). This suggests that biomass torrefied

under proper conditions could give a higher gasification efficiency as higher surface area and porosity can increase the contact area between the fuel and gasification medium and accelerate the rate of reactions and mass transfer.

Fisher et al., (2012) reported that the combustion and gasification (using  $\text{H}_2\text{O}/\text{N}_2$  as the gasifying medium) reactivities of chars produced from torrefied willow is lower than those of raw willow. Min et al., (2011) gasified pyrolysed agricultural waste with a mixture of  $\text{CO}_2/\text{N}_2$  and reported that the overall char reactivity decreased with an increase of pyrolysis temperature. It should be noted that both studies were carried out using a thermo-gravimetric analyser. The reactivity of the samples was evaluated by the mass loss rate. But the mass loss rate is not always identical to the reactivity of feedstock from the perspective of reaction mechanism of gasification, for example the release of volatile matters.

Extensive research has been carried out to investigate the influence of torrefaction on the fuel properties of biomass and its potential effects on the gasification process, for example Prins et al., (2006) compared three gasification scenarios from the point view of mass and energy balances and reported that torrefaction prior to gasification is a promising method to achieve more efficient gasification of wood. Publications reporting gasification of torrefied biomass in actual gasifiers are rare. Couhert et al., (2009) gasified raw and torrefied wood samples in a high temperature entrained flow reactor with 20 vol. % steam in  $\text{N}_2$ . They concluded that torrefied wood produced more  $\text{H}_2$  and CO than raw wood at 1400 °C. Chen and co-workers (2013) numerically compared the performance of raw and torrefied bamboo and high volatile bituminous coal in an entrained flow gasifier using  $\text{O}_2$  as the gasification medium. It was found that torrefaction made the gasification behaviour of biomass approach that of coal; the

torrefaction was able to facilitate syngas formation from biomass gasification. Moreover, the cold gasification efficiency of torrefied bamboo was improved by 88% compared to raw bamboo under the optimum conditions. Stelt et al., (2011) reported that torrefaction is the most cost-effective and environmental friendly technology for the Biomass-to-Liquid (BTL) plant located in the Netherlands with a capacity of 1000 MWth synthesis gas.

As there are relatively few reports in the scientific literature with detailed investigation of process conditions for the gasification of torrefied MxG, the objectives of this study are to: (1) investigate the influence of main gasification parameters, such as equivalence ratio (ER) and temperature, on the performance of torrefied MxG in an air-blown bubbling fluidized bed gasifier; (2) propose the optimal conditions for the gasification of torrefied MxG; (3) preliminary comparison of the gasification characteristics of raw and torrefied MxG from the point view of gasification efficiency.

## **2. Materials and methods**

### **2.1 Biomass torrefaction, characterization and bed material**

The biomass feedstock used in this study, *Miscanthus x giganteus* (MxG) pellets, were supplied by JHM Crops, Ireland. which were crushed to less than 5 mm. It was torrefied using a batch reactor at 250 °C at 20 °C/min. The average yield for all batches was 76 wt. %. The torrefied MxG was milled in a ball mill prior to characterisation and testing.

The properties of the raw and torrefied MxG are presented in Table 1. The methods can be found elsewhere (Xue et al., 2014). Ash content and volatile matter content are

expressed on an as received basis. Fixed carbon content was calculated according to Basu (2010a). The bed material used in this study was non-calcined olivine (250 - 500  $\mu\text{m}$ ) supplied by Eurogrit, the Netherlands with bulk and absolute densities of 1573.8 and 3171.5  $\text{kg/m}^3$  respectively.

## 2.2 Experimental gasifier

The experiments were carried out on an air-blown bubbling fluidized bed gasifier at Energy Research Centre of Netherlands (ECN). The reactor consisted of a bed section (500 mm high and 74 mm internal diameter (ID)) and a freeboard section (600 mm high and an ID of 108 mm). Both sections were externally heated. The gasification medium (air and  $\text{N}_2$ ) were introduced through a gas distributor at the base of the gasifier. The solid fuel was fed 50 mm above the gas distributor by means of a feeding screw. The detailed information can be found in the report published by Paasen et al. (2006).

## 2.3 Experiment procedure

### 2.3.1 Gasification procedure

For each experiment, the gasifier was charged with 1.15 kg (725 ml) of raw olivine and heated to a set point 50  $^{\circ}\text{C}$  lower than the desired gasification temperature before feeding commenced. The feeding rate of biomass was constant ( $\sim 580 \pm 11$  g/h) for all the gasification tests conducted on the torrefied material. The equivalence ratio (ER) was defined and calculated using the methods described by Thomas and Allan (1993). In order to keep the fluidization conditions the same for all tests, a stream of pure  $\text{N}_2$  was added to keep a constant gasification medium flow when the ER was varied. It

should be noted that only the N<sub>2</sub> from air was accounted in the product gas. The superficial fluidisation velocity was calculated according to Siedlecki et al. (2011).

Once steady state was achieved, tar (solid phase adsorption (SPA)) and moisture samples were taken (see section 2.3.3). The composition of the produced gas was measured continuously during experiments (section 2.3.2).

Before each experiment was finished, a known flow rate of Ar (0.4 dm<sup>3</sup>/min) was introduced into the gasifier together with the gasification agent (air and N<sub>2</sub> mixture) to calculate the flow rate of the product gas (see section 2.3.2). The flow of Ar was maintained for at least 20 minute after its concentration measurement had stabilized. The flow rate of product gas was calculated according to equation (1) using the concentration of the internal standard gas-Ar. Where,  $\beta_{\text{product gas}}$ ,  $\beta_{\text{air}}$ , and Ar represent the flow rate of product gas (dm<sup>3</sup>/min) and air (dm<sup>3</sup>/min), the concentration of Ar (%) in the product gas after the additional Ar was introduced.

$$\beta_{\text{product gas}} = \left( \frac{\beta_{\text{air}} \times 0.9\% + 0.4}{\text{Ar}} \right) \times 100 - 0.4 \quad (1)$$

The rate of biomass feeding and generation of elutriated char in the cyclone were calculated as average values during the gasification period.

### 2.3.2 Product gas analysis

The composition of the filtered dry product gas, which passed through the cyclone and both the hot and cold filters was determined using a Varian, CP4900 Micro-GC that had been calibrated using five different gas mixtures (step concentration) prior to the experiments. An ABB gas analyser was used to determine the H<sub>2</sub> and O<sub>2</sub> content.

The gas analysis was carried out according to the method described by Paasen et al. (2006).

### **2.3.3 Moisture quantification**

A simple method was used to determine the moisture content of the product gas in an N<sub>2</sub> containing gas. Specifically, 100 ml of hot gas was drawn through 1 g of phosphorous pentoxide (P<sub>2</sub>O<sub>5</sub>) at 50 ml/min allowing the gas to cool down to room temperature (about 25°C) and achieve a full adsorption. The moisture content was calculated as the mass gained after adsorption. Although most of the tar compounds will condense and be adsorbed, the mass of moisture in the product gas is expected to be an order of magnitude higher than tar which makes this method sufficiently accurate for the present purpose.

### **2.3.4 Tar analysis**

The tar samples were taken by means of SPA according to the method described by Osipovs, (2009). The tar compounds were extracted by dichloromethane (DCM) and Phenol-d6 and Naphthalene-d8 were added as internal standards. An Agilent 7890A Gas Chromatograph (GC) coupled with a HP-5MS capillary column (5% diphenyl 95% dimethylpolysiloxane) and triple-axis MSD 5975C was employed to identify and quantitatively analyse GC detectable tar compounds. In order to increase the accuracy, the phenolic compounds quantified by Phenol-d6, all the rest were quantified by Naphthalene-d8. At least two duplicates were used for all the samples.

### **2.3.5 Gasification efficiency**



The higher heating value of product gas,  $\text{HHV}_P$  ( $\text{MJ}/\text{m}^3$ ) and cold gas efficiency (CGE),  $\eta_{\text{CG}}\%$  were determined according to Basu, (2010b) while the carbon and hydrogen conversion efficiencies,  $\eta_c$  (%) and  $\eta_H$  (%) respectively were determined according to Basu, (2006):

### 2.3.6 Mass balance and distribution calculations.

The mass balance is defined as the ratio of mass output from the gasifier (product gas, elutriated char, tar and moisture) to input to the gasifier (biomass and air) in order to evaluate the overall fuel utilization efficiency of a gasification process or apparatus. It should be noted that it was assumed that there was no biochar accumulation within the gasifier **and** the mass of biochar in the gasifier was constant, so there was no need to consider how much char remained in the bed after gasification. This assumption has been used by other researchers Siedlecki et al., (2009).

The mass input from the biomass was converted into the input of carbon (C), hydrogen (H), nitrogen (N) and oxygen (O) (kg/h) on a moisture and ash free basis (corresponding to the elemental analysis). The moisture from the biomass was divided into the H and O input. The mass flow of air was calculated from its volumetric flow rate ( $\text{dm}^3/\text{min}$ ) and density. The mass flow of the gas species in the air ( $\text{N}_2$ ,  $\text{O}_2$  and  $\text{CO}_2$ ) was calculated according to their mole fractions and converted to CHNO flow (kg/h). The sum of all elements represented the overall mass input. The mass flow rate of each gas species (kg/h) in the product gas was calculated as the product of its density ( $\text{kg}/\text{m}^3$ ), concentration and yield of the product gas ( $\text{m}^3/\text{h}$ ). Char elutriation (collected from the cyclone) was estimated as the average value over the entire period of gasification. Char samples collected after gasification were analyzed for moisture, ash and elemental composition. The moisture content of product gas was taken from

section 2.3.3. Each stream was included in the CHNO output. The sum of CHNO from all the streams was the overall mass output. It should be noted that the mass of tar was included as moisture according to the assumption made in the Section 2.3.3.

### **3. Results and discussion**

#### **3.1 Differential thermo-gravimetric (DTG) analysis of raw and torrefied miscanthus**

Lignocellulosic biomass consists of three main constituents: cellulose, hemicellulose and lignin. Because of intrinsic differences in the chemical structure of these constituents, they can be distinguished and identified using thermo-gravimetric analysis on the basis of the differing pyrolytic reactivity they exhibit (Chen & Kuo, 2010; Chen, 2011b).

In order to understand the influence of torrefaction on the lignocellulose composition of the MxG, both raw and torrefied MxG were subjected to the thermo-gravimetric analysis (TGA). Approximately 5 mg of sample was used for each experiment and the sample was heated at a rate of 10 °C/min from room temperature to 110 °C and it was maintained for 10 minutes to evaporate moisture. Subsequently, the temperature was increased to 900 °C at the same heating rate and it was maintained for 10 minutes. The entire process was performed under a N<sub>2</sub> purge flow. Figure 1 shows the difference in the chemical nature of the torrefied and raw MxG. The thermogram can be divided into three regions. From room temperature to 105 °C, the peaks originated from the evaporation of moisture in the biomass. As raw MxG has higher moisture content than torrefied MxG a higher peak ought to be expected. In the

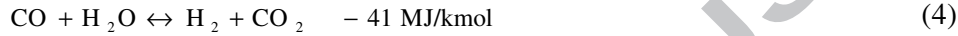
temperature range of 105 and 380 °C, a shoulder peak at ~300 °C (partially merged) was observed in addition to the main peak at 340 °C for raw MxG. These peaks reflect the decomposition of hemi-cellulose and cellulose respectively. In the case of torrefied MxG, only a single large peak appeared at ~343 °C representing cellulose degradation. This suggests that most of the hemi-celluloses were decomposed during the process of torrefaction. The proportion of cellulose was augmented compared to that of raw miscanthus. In the third region between 380 and 900 °C, the curves slowly approached the x-axis which is the typical decomposition pattern of lignin. It should be noted that the peak of torrefied miscanthus was higher in the range of 380 and 480 °C indicating higher lignin content.

### **3.2 Effect of equivalent ratio (ER)**

#### **3.2.1 Effect of ER on the composition of product gas**

ER is an important gasification parameter having a significant influence on the composition of the product gas from air-blown biomass gasification as well as overall gasification efficiency. In this study the effect of ER was investigated between 0.18 to 0.26 (at a constant temperature (800 °C) and biomass feeding rate). Different ERs were obtained by changing the proportion of air and N<sub>2</sub> flows, but the total amount of fluidization medium remained the same at 12.3 dm<sup>3</sup>/min. The change of gas composition in the product gas as a function of ER is plotted in Figure 2 (a). It can be seen that the concentration of H<sub>2</sub> and CO decreased from 10.2 to 6.4% and 17.0 to 14.5% respectively with increasing ER. The main reason for the decrease of H<sub>2</sub> and CO is the increased stoichiometric O<sub>2</sub> supply which gives rise to oxidation reactions of (5), (9) and (10), listed together with the other main reactions (2)-(10) occurring during

gasification. This was verified by the increased concentration of CO<sub>2</sub> in the product gas which increased from 13.7 to 15.0%. Similar results were reported by other researchers (Karatas et al., 2012; Manyà et al., 2006). Additionally, the N<sub>2</sub> content in the product gas was increased from 51.59 to 57.33%, due to the diluting effect of higher ER. The concentration of light hydrocarbons decreased by 10 to 20% over the ER range.



### 3.2.2 Effect of ER on the performance of gasification

Figure 2 (b) illustrates the effect of ER on the higher heating value (HHV), gas yield, carbon/hydrogen conversion and cold gas efficiency. It shows that the HHV decreased with increasing ER up to a value of 0.23, due to the reduced concentration of CO and H<sub>2</sub> which were consumed by oxidation reactions (section 3.1.1) and the diluting effect of increased N<sub>2</sub> in the product. Similar results have been reported by other researchers (Arena et al., 2010; Kim et al., 2013). However, when the ER was increased from 0.23 to 0.26 no further decrease in HHV was observed. This may be

due to minor increases in the concentration of light hydrocarbons probably induced by tar cracking. Although the increase in concentration of ethylene and methane was changed slightly, they have much higher HHV than that of CO and H<sub>2</sub>. Thus, a slight change in their concentrations can have a significant influence on the value of HHV. The cold gasification efficiency, which were in the range of 54.9 to 62.7%.

The gas yield is defined as the flow rate of dry gas produced to mass flow rate of dry ash free feedstock. As can be seen in Figure 2(b), the gas yield increased with ER. While the measured data showed an obvious trend, the actual values may be subject to some determinate source of error which was not identified at the time of experiments. With regards to carbon and hydrogen conversion, they were in the range of 73.5 to 82.5% and 54.9 to 62.7% respectively. The obvious increases at the highest ER 0.26 could be the result of tar cracking. Chen and co-workers reported similar results (Chen et al., 2013).

### 3.2.3 Effect of ER on the absolute gas yield

The absolute yield of each gas species on the basis of unit mass of biomass ( $\text{m}^3/\text{kg}_{\text{biomass}}$ ) is of great importance for its potential utilization, and consequently it is presented in Figure 2 (c) and (d). It was observed that the absolute yield of H<sub>2</sub> decreased progressively from 0.184 to 0.138  $\text{m}^3/\text{kg}_{\text{biomass}}$  with the increase of ER. As expected, the yield of CO<sub>2</sub> increased with ER and had the highest value in the product gas at an ER of 0.26. On the other hand, the yield of light hydrocarbons was an order of magnitude lower than that of main gas species and remained relatively constant. Amongst the hydrocarbons, CH<sub>4</sub> was the most abundant component and followed by C<sub>2</sub>H<sub>4</sub>. The contents of ethane, acetylene, benzene and toluene were very low and were

not measurably affected by a change in ER. It can be concluded that the optimal ER for torrefied MxG was 0.21.

### 3.3 Effect of temperature

#### 3.3.1 Effect of temperature on the composition of product gas

Since the temperature is another critical parameter which potentially has a significant effect on the gasification process, its influence was investigated between 660 and 850 °C for the optimal ER 0.21. It should be noted that the slight fluctuation of ER for the temperature trials resulted from the minor changes in the biomass feeding. Figure 3 (a) presents the changes in gas composition of the product gas as a function of gasification temperature. It was observed that the concentration of H<sub>2</sub> increased from 5.5 to 10.4% when the temperature in the bed was raised from 660 to 850 °C. It may be the overall result of the promoted water gas shift (WGS) reaction (4), dry (7) and wet (6) tar reforming and the water gas reaction (10) (Mayerhofer et al., 2012; Mohammed et al., 2011). The content of CO rose from 12.4 to 17.8% which could be due to the improved Boudouard reaction (2), carbon partial oxidation (3) and water gas reaction (8). This trend is in agreement with results published by other researchers (Lahijani & Zainal, 2011; Mayerhofer et al., 2012; Mohammed et al., 2011). On the other hand, the content of CO<sub>2</sub> fell from 16.7 to 12.9% in the same temperature range. This observation differs from a study by Lahijani and co-workers' (2011). But Mohammed *et al.*, (2011) reported a similar trend observing a decrease of CO<sub>2</sub> content with temperature for the gasification of palm empty fruit bunches. The suggested reason is that the CO<sub>2</sub> was consumed, as mentioned, by the Boudouard (2) and tar dry reforming (7) reactions. In addition, at elevated temperature in the gasifier the exothermic water

gas shift reaction (4) was shifted to the  $\text{H}_2\text{O}$  and  $\text{CO}$  side which lowered the yield of  $\text{CO}_2$  (Figure 3c). In the case of  $\text{N}_2$ , it was fell moderately from 59.1 to 51.7% due to improved gasification and tar cracking reactions. The trends of light hydrocarbons did not show obvious variations in Figure 3 (a), but their absolute yield experienced significant changes that will be discussed in Section 3.3.3.

### 3.3.2 Effect of temperature on the performance of gasification

The HHV, gas yield and gasification performance (carbon/hydrogen conversion and cold gas efficiency) is presented in Figure 3 (b). It can be seen that the overall influence of temperature on the gasification results and performance is opposite to that of ER (Figure 2 (b)). It was observed that the HHV experienced a slight increase from 660 to 700 °C, followed by a significant rise between 700 and 850 °C and peaked at 7.05 MJ/m<sup>3</sup> at the highest experimental temperature. The gas yield increased by 23.5% from 1.7 to 2.1 m<sup>3</sup>/kg<sub>biomass</sub> which resulted from the promoted biomass pyrolysis and gasification, tar cracking/reforming and endothermic char gasification reactions (Pinto et al., 2003). Accordingly, the cold gasification efficiency was lifted by 70%. Meanwhile, the conversion of carbon was improved by 34% and hydrogen conversion by 69% with the increase of gasification temperature.

### 3.3.3 Effect of temperature on the absolute gas yield

The absolute yield of both major and minor gas species is plotted in the Figure 3 (c) and (d) respectively. In the Figure 3 (c), it is seen that the yield of  $\text{CO}$  increased substantially with temperature. It started to increase from 0.210 m<sup>3</sup>/kg<sub>biomass</sub> at 660 °C, exceeded the yield of  $\text{CO}_2$  at 800 °C and reached its highest value at 850 °C with 0.365 m<sup>3</sup>/kg<sub>biomass</sub>. On the other hand, the highest yield of  $\text{CO}_2$  was observed at 700 °C and

then it decreased progressively with increase in gasification temperature. Regarding  $H_2$ , its yield more than doubled from  $0.092 \text{ m}^3/\text{kg}_{\text{biomass}}$  at  $660^\circ\text{C}$  to  $0.214 \text{ m}^3/\text{kg}_{\text{biomass}}$  at  $850^\circ\text{C}$ . As presented in Figure 3 (d), the yield of  $C_2H_4$ ,  $C_2H_2$  and benzene increased with increasing process temperature which is in agreement with previous work on thermal decomposition of hydrocarbons (Taralas et al., 1991). But that of  $C_2H_6$  was decreased. The yield of light hydrocarbons from the temperature tests was of the similar magnitude to that of ER tests (Figure 2 (c) and (d)), but fluctuated more significantly when the temperature changed which implies that the influence of temperature is more significant than that of ER on the yield of light hydrocarbons. It should be noted that agglomeration was not observed for any of the temperature tests except test 8 in which the bed was defluidized approximately 50 minutes after the temperature reached  $850^\circ\text{C}$ . Considering the sustainability of gasification process, quality of product gas, overall energy yield and gasification efficiency of all the temperature trials, the temperature of  $800^\circ\text{C}$  was nominated as the optimum temperature. Thus the optimal condition for the gasification of torrefied M×G was 0.21 ER and  $800^\circ\text{C}$  temperatures (test 2).

### 3.4 Comparison of raw and torrefied M×G

An experiment was initially designed to gasify the raw M×G at the optimal conditions (ER of 0.21 and  $800^\circ\text{C}$ ) resulting from the tests of torrefied M×G, so as to compare the performance of the two different feedstocks. But the actual ER calculated after the test (based on the actual biomass feeding rate) for the experiment with raw M×G was found to be 0.32 which deviated significantly from the set point (0.21) because the biomass feeding rate had changed. The altered ER made it impossible to



draw conclusions in the manner planned. The detailed investigation will be conducted later on.

### 3.5 Mass balance and distribution

The mass balance and mass distribution for all the gasification tests conducted in this study is presented in Table 3. As can be seen, the mass balances for all the cases were higher than 100% indicating the output was higher than the input, deviation (above 100%) was calculated and is presented above each column. The possible reasons of discrepancies are that: (1) many different kinds of analysis were carried out to provide data for the mass balance calculation (measurement errors); (2) the errors which are not possible to be eliminated completely can be accumulated and exaggerated after multiple calculations; (3) there was a systematic error in the gas yield determination, which would also lead to a systematic error in the cold gas efficiency; (4) tar compounds could be adsorbed by  $P_2O_5$  (section 2.3.3) and calculated as moisture. Since the scale of errors is relatively small, the results are considered to be acceptable. The discussion of mass balance for gasification process is scarce in the scientific literatures. The deviation of results in mass balance were only found in a few publications (Siedlecki & de Jong, 2011; Siedlecki et al., 2009).

It is seen that both torrefied and raw MxG were mainly converted into permanent gases. It should be noted that the overall performance of torrefied and raw MxG (test 2 and 9), the mass of product gas from both tests was almost the same; however, the amount of elutriated char and moisture fraction were 5.8 and 2.2 times lower for the raw biomass (test 9).

#### **4. Conclusion**

Raw and torrefied M×G were gasified in an air-blown BFB gasifier to investigate the influence of ER and temperature on the gasification performance. The gasification of torrefied M×G yields 1.7-2.2 m<sup>3</sup>/kg<sub>biomass</sub> product gas with a LHV between 4.7 and 6.6 MJ/m<sup>3</sup>. The gas yield increased with gasification temperature. The conversion efficiency increased with temperature but fluctuated when ER was elevated. Preliminary comparison suggests that torrefied might be superior to raw M×G from the perspective of gasification efficiency. Torrefaction prior to gasification is a promising route for the energy production via air-blow BFB gasification.

#### **Acknowledgements**

The financial support provided by the EU project Biomass Research Infrastructure for Sharing Knowledge (BRISK) and Enterprise Ireland Competence Centre for Biorefining & Biofuels as research grants (CC/2009/1305A) are gratefully acknowledged. The authors wish to thank the Energy Research Centre of Netherlands (ECN) for the technical support and cooperation. Gang Xue acknowledges the postgraduate research scholarship received from Chinese Scholarship Council. Stephen Dooley is supported by Science Foundation Ireland under Grant No: 799 06/CP/E007.

#### **Supplementary data**

Supplementary data associated with this article can be found, in the online version.

#### **Reference**

1. Arena, U., Zaccariello, L., Mastellone, M.L. 2010. Fluidized bed gasification of waste-derived fuels. *Waste Manag.* 30, 1212-1219.
2. Basu, P. 2010a. *Biomass Gasification and Pyrolysis - Practical Design and Theory*, Academic Press. Boston, **p. 50**.
3. Basu, P. 2006. Bubbling Fluidized Bed Boiler. in: *Combustion and Gasification in Fluidized Beds*, CRC Press, pp. 211-252.
4. Basu, P. 2010b. Chapter 6 - Design of Biomass Gasifiers. in: *Biomass Gasification and Pyrolysis*, Academic Press. Boston, pp. 167-228.
5. Bridgeman, T.G., Jones, J.M., Shield, I., Williams, P.T. 2008. Torrefaction of reed canary grass, wheat straw and willow to enhance solid fuel qualities and combustion properties. *Fuel*, 87, 844-856.
6. Chen, Q., Zhou, J.S., Liu, B.J., Mei, Q.F., Luo, Z.Y. 2011a. Influence of torrefaction pretreatment on biomass gasification technology. *Chinese Sci Bull*, 56(14), 1449-1456.
7. Chen, W.-H., Chen, C.-J., Hung, C.-I., Shen, C.-H., Hsu, H.-W. 2013. A comparison of gasification phenomena among raw biomass, torrefied biomass and coal in an entrained-flow reactor. *Appl Energy*, 112, 421-430.
8. Chen, W.-H., Kuo, P.-C. 2010. A study on torrefaction of various biomass materials and its impact on lignocellulosic structure simulated by a thermogravimetry. *Energy*, 35, 2580-2586.
9. Chen, W.-H., Kuo, P.-C. 2011b. Torrefaction and co-torrefaction characterization of hemicellulose, cellulose and lignin as well as torrefaction of some basic constituents in biomass. *Energy*, 36, 803-811.
10. Couhert, C., Salvador, S., Commandré, J.M. 2009. Impact of torrefaction on syngas production from wood. *Fuel*, 88, 2286-2290.

11. Fisher, E.M., Dupont, C., Darvell, L.I., Commandre, J.M., Saddawi, A., Jones, J.M., Grateau, M., Nocquet, T., Salvador, S. 2012. Combustion and gasification characteristics of chars from raw and torrefied biomass. *Bioresource Technol*, 119, 157-165.
12. Karatas, H., Olgun, H., Akgun, F. 2012. Experimental results of gasification of waste tire with air&CO<sub>2</sub>, air&steam and steam in a bubbling fluidized bed gasifier. *Fuel Processing Technol*, 102, 166-174.
13. Kim, Y.D., Yang, C.W., Kim, B.J., Kim, K.S., Lee, J.W., Moon, J.H., Yang, W., Yu, T.U., Lee, U.D. 2013. Air-blown gasification of woody biomass in a bubbling fluidized bed gasifier. *Appl Energy*, 112, 414-420.
14. Lahijani, P., Zainal, Z.A. 2011. Gasification of palm empty fruit bunch in a bubbling fluidized bed: A performance and agglomeration study. *Bioresource Technol*, 102, 2068-2076.
15. Mansaray, K.G., Ghaly, A.E., Al-Taweel, A.M., Hamdullahpur, F., Ugursal, V.I. 1999. Air gasification of rice husk in a dual distributor type fluidized bed gasifier. *Biomass Bioenergy*, 17, 315-332.
16. Manyà, J.J., Sánchez, J.L., Ábrego, J., Gonzalo, A., Arauzo, J. 2006. Influence of gas residence time and air ratio on the air gasification of dried sewage sludge in a bubbling fluidised bed. *Fuel*, 85, 2027-2033.
17. Mayerhofer, M., Mitsakis, P., Meng, X., de Jong, W., Spliethoff, H., Gaderer, M. 2012. Influence of pressure, temperature and steam on tar and gas in allothermal fluidized bed gasification. *Fuel*, 99, 204-209.

18. Min, F., Zhang, M., Zhang, Y., Cao, Y., Pan, W.-P. 2011. An experimental investigation into the gasification reactivity and structure of agricultural waste chars. *J Anal Appl Pyrol*, 92, 250-257.
19. Mohammed, M.A.A., Salmiaton, A., Wan Azlina, W.A.K.G., Mohammad Amran, M.S., Fakhru'l-Razi, A. 2011. Air gasification of empty fruit bunch for hydrogen-rich gas production in a fluidized-bed reactor. *Energy Convers Manag*, 52, 1555-1561.
20. Osipovs, S. 2009. Use of two different adsorbents for sampling tar in gas obtained from peat gasification. *International Journal of Environmental Analytical Chemistry*, 89, 871-880.
21. Paasen, S.V.B.v., Cieplik, M.K., Phokawat, N.P. 2006. Gasification of Non-woody Biomass--Economic and Technical Perspectives of Chlorine and Sulphur Removal from Product Gas (Non-confidential version). ECN.
22. Phanphanich, M., Mani, S. 2011. Impact of torrefaction on the grindability and fuel characteristics of forest biomass. *Bioresource Technol*, 102, 1246-1253.
23. Pinto, F., Franco, C., André, R.N., Tavares, C., Dias, M., Gulyurtlu, I., Cabrita, I. 2003. Effect of experimental conditions on co-gasification of coal, biomass and plastics wastes with air/steam mixtures in a fluidized bed system. *Fuel*, 82, 1967-1976.
24. Prins, M.J., Ptasinski, K.J., Janssen, F.J.J.G. 2006. More efficient biomass gasification via torrefaction. *Energy*, 31, 3458-3470.
25. Siedlecki, M., de Jong, W. 2011. Biomass gasification as the first hot step in clean syngas production process – gas quality optimization and primary tar reduction measures in a 100 kW thermal input steam–oxygen blown CFB gasifier. *Biomass Bioenergy*, 35, Supplement 1, S40-S62.

26. Siedlecki, M., De Jong, W., Verkooijen, A.H.M. 2011. Fluidized Bed Gasification as a Mature And Reliable Technology for the Production of Bio-Syngas and Applied in the Production of Liquid Transportation Fuels—A Review. *Energies*, 4, 389-434.
27. Siedlecki, M., Nieuwstraten, R., Simeone, E., de Jong, W., Verkooijen, A.H.M. 2009. Effect of Magnesite as Bed Material in a 100 kWth Steam–Oxygen Blown Circulating Fluidized-Bed Biomass Gasifier on Gas Composition and Tar Formation. *Energy Fuels*, 23, 5643-5654.
28. Stelt, M.J.C.v.d., Gerhauser, H., Kiel, J.H.A., Ptasiński, K.J. 2011. Biomass upgrading by torrefaction for the production of biofuels: A review. *Biomass Bioenergy*, 35, 3748-3762.
29. Sweeney, D.J. 2012. Performance of a pilot scale, steam blown, pressurized fluidized bed biomass gasifier. in: Department of Mechanical Engineering Vol. Doctor, University of Utah, p. 94.
30. Taralas, G., Vassilatos, V., Sjöström, K., Delgado, J. 1991. Thermal and catalytic cracking of n-Heptane in presence of CaO, MgO and Calcined Dolomites. *Can J Chem Eng*, 69, 1413-1419.
31. Thomas, D.E., Allan, M. 1993. Applied Thermodynamics for Engineering Technologists 5th ed. Pearson Education Limited, pp. 183-185.
32. Van der Stelt, M.J.C., Gerhauser, H., Kiel, J.H.A., Ptasiński, K.J. 2011. Biomass upgrading by torrefaction for the production of biofuels: A review. *Biomass Bioenergy*, 35, 3748-3762.
33. Xiao, R., Jin, B., Zhou, H., Zhong, Z., Zhang, M. 2007. Air gasification of polypropylene plastic waste in fluidized bed gasifier. *Energy Convers Manage*, 48, 778-786.

34. Xue, G., Kwapinska, M., Horvat, A., Li, Z., Dooley, S., Kwapinski, W., Leahy, J.J.  
2014. Gasification of *Miscanthus x giganteus* in an air-blown bubbling fluidized bed: a  
preliminary study of performance and agglomeration. Energy & Fuels. DOI:  
10.1021/ef4022152

**List of Figures:**

**Figure 1.** DTG distribution of raw and torrefied miscanthus

**Figure 2.** The effect of ER on (a) the gas composition of the product gas; (b) the performance of gasification; (c) the absolute yield of main gas species; (d) the absolute yield of the light hydrocarbon gas species in the product gas.

**Figure 3.** The effect of temperature on (a) the composition of the product gas; (b) the performance of gasification; (c) the absolute yield of main gas species; (d) the absolute yield of the light hydrocarbon gas species in the product gas.



**List of Tables:**

**Table 1.** Properties of raw and torrefied MxG.

**Table 2.** Moisture content in the product gas from gasification tests.

**Table 3.** Mass balance and distribution (wt. %) of gasification tests

**List of supplements:**

**Supplementary material 1.** The main parameters used in MxG gasification tests and the corresponding results.

**Supplementary material 2.** Standard deviation of gas composition measurement

ACCEPTED MANUSCRIPT

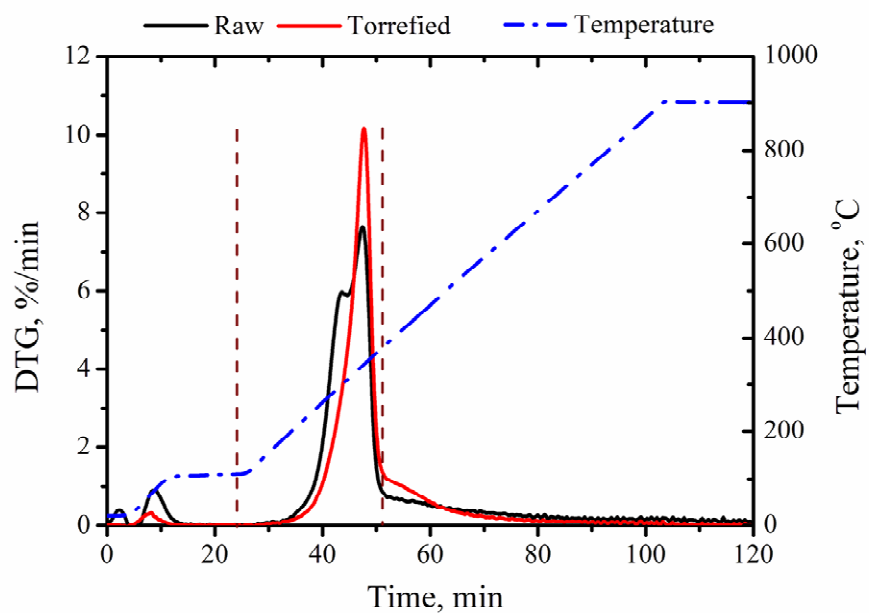


Figure 1.

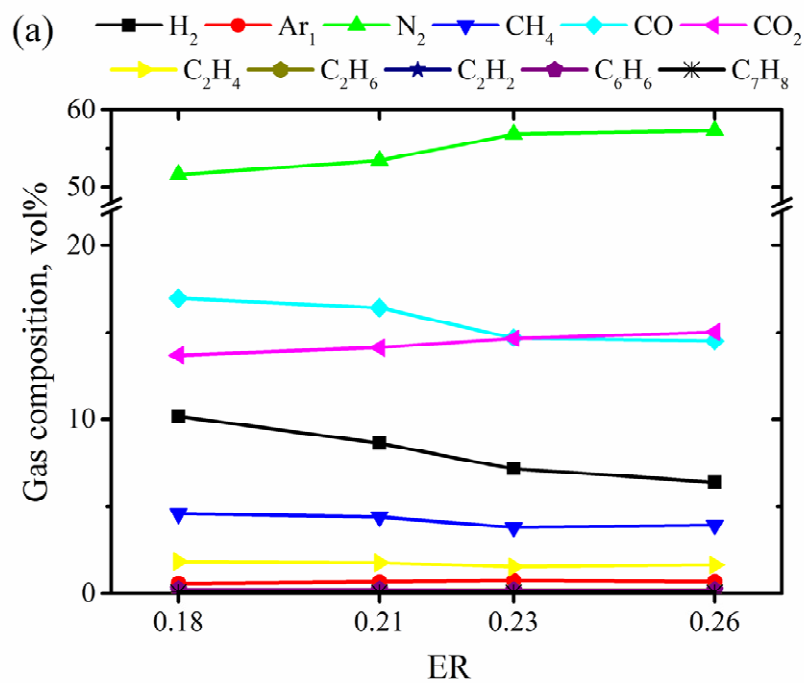


Figure 2 (a)

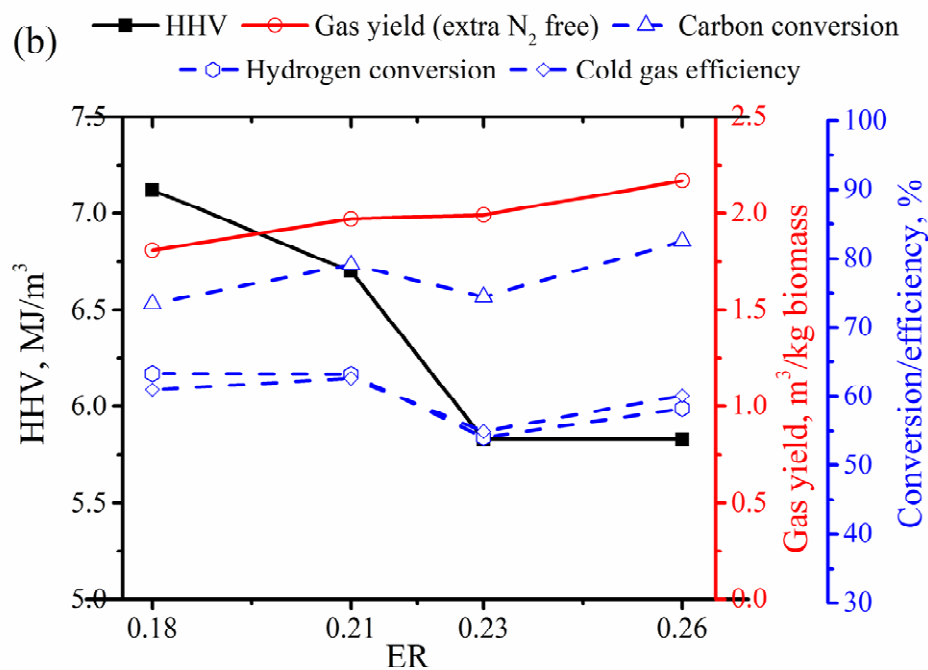


Figure 2 (b)

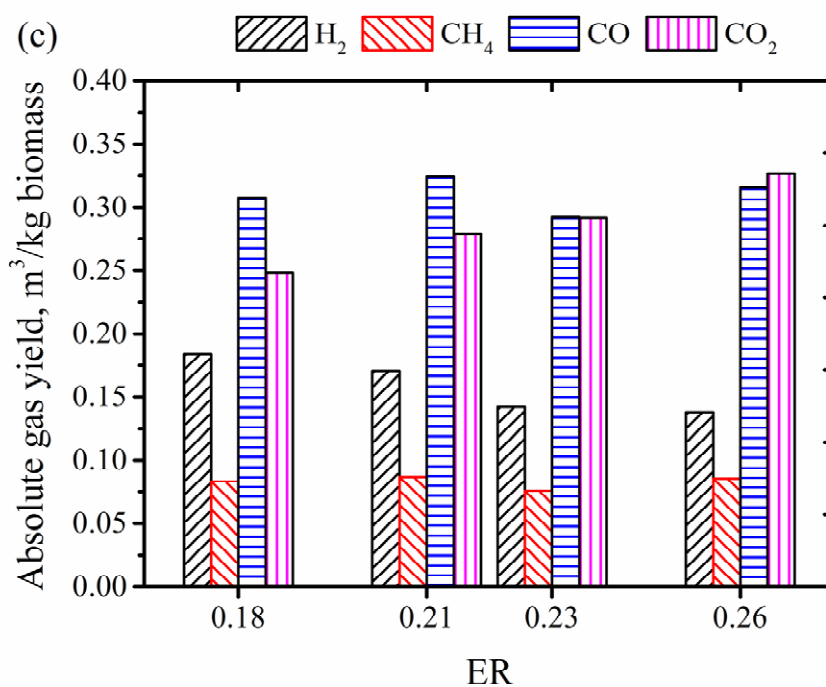


Figure 2 (c)

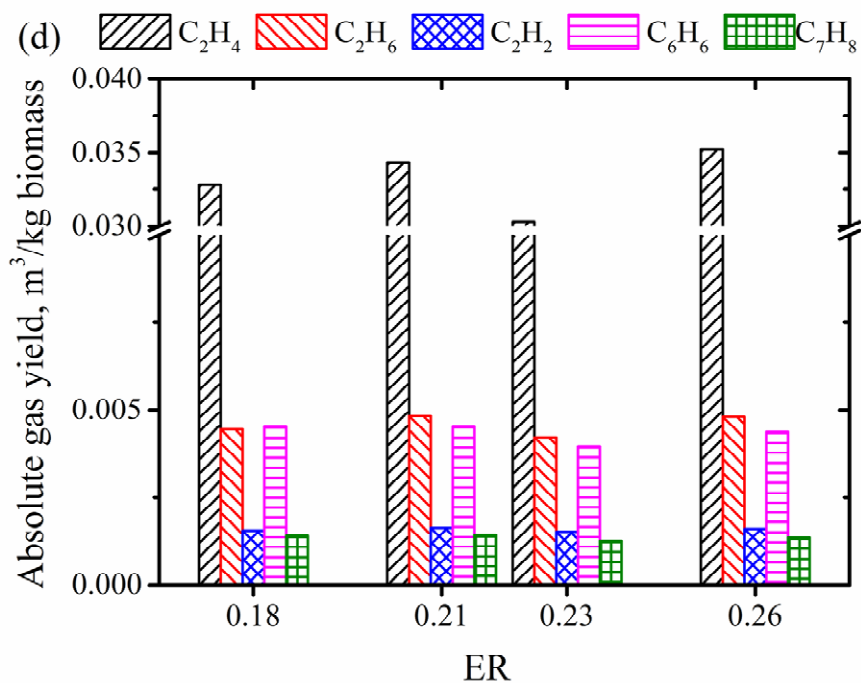


Figure 2 (d)

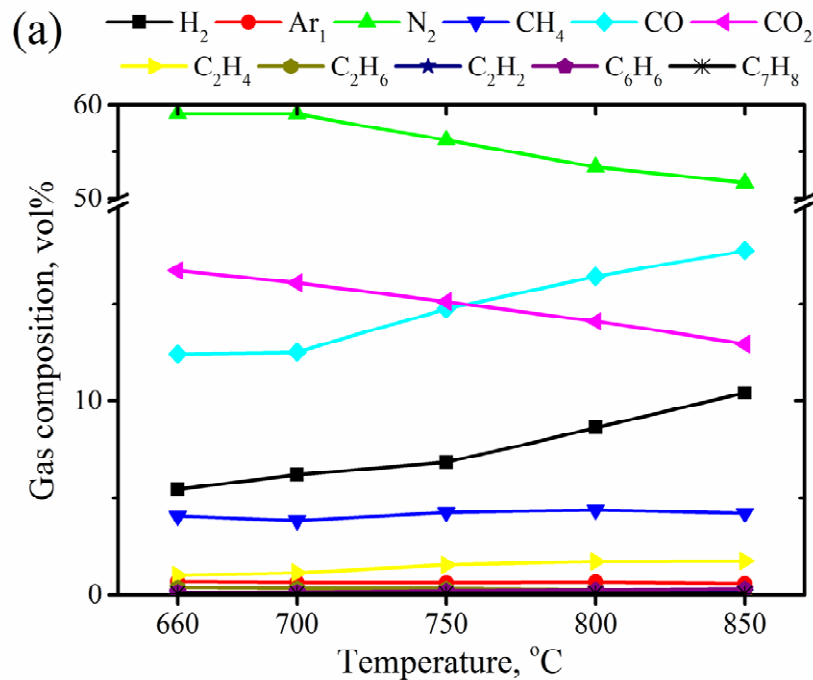


Figure 3 (a)

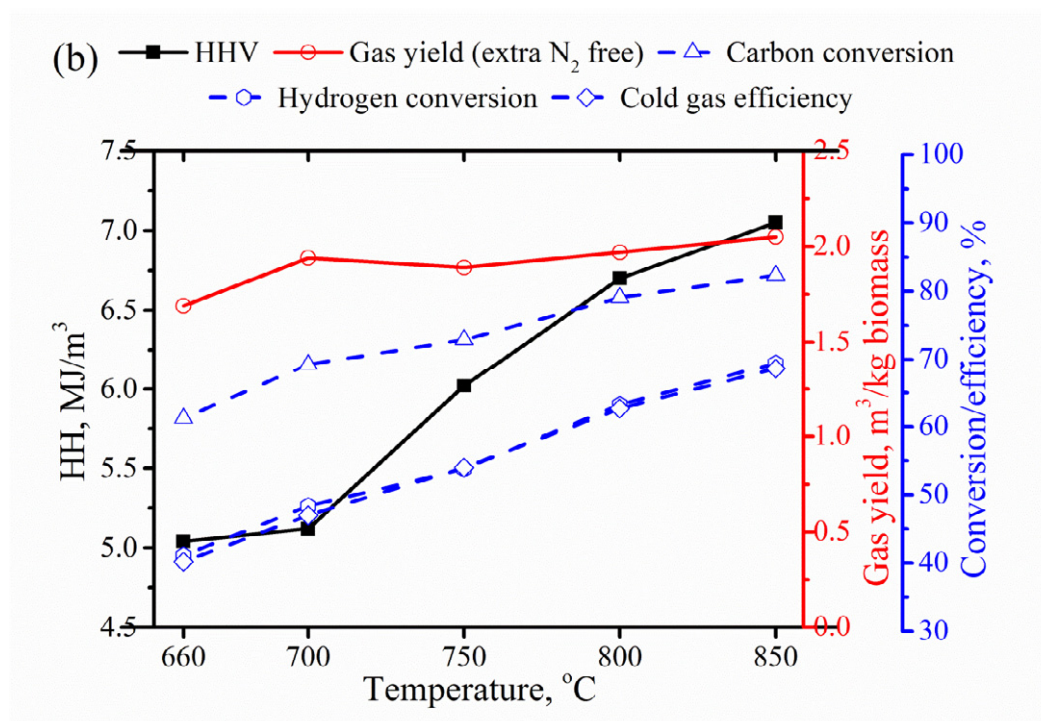


Figure 3 (b)

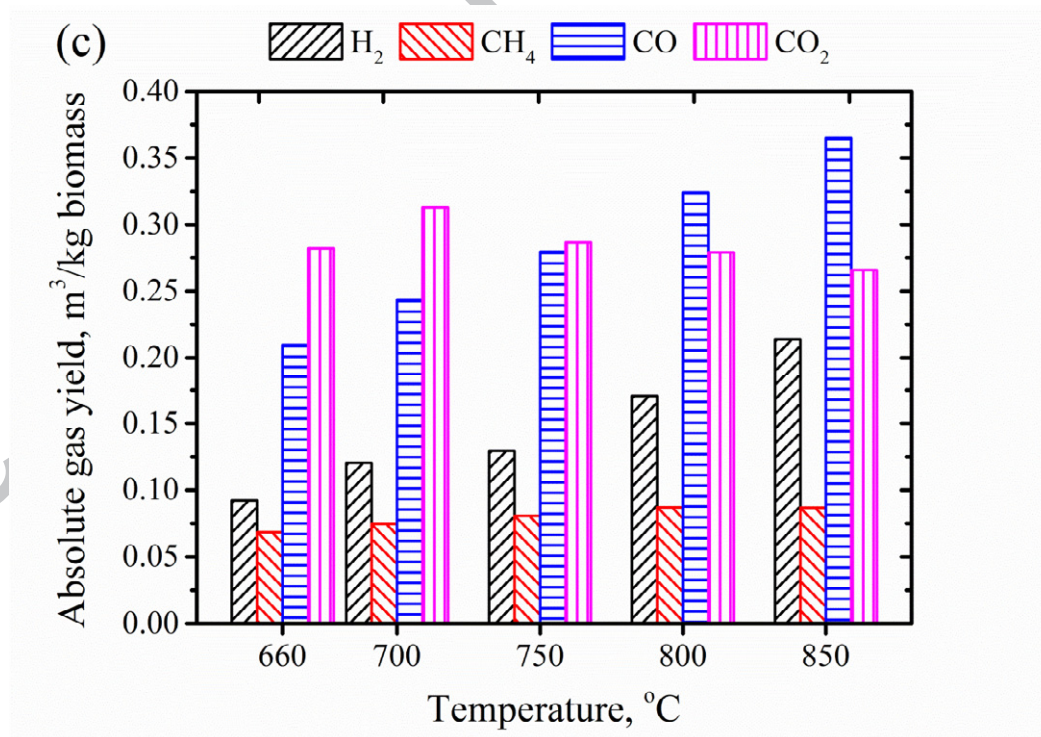


Figure 3 (c)

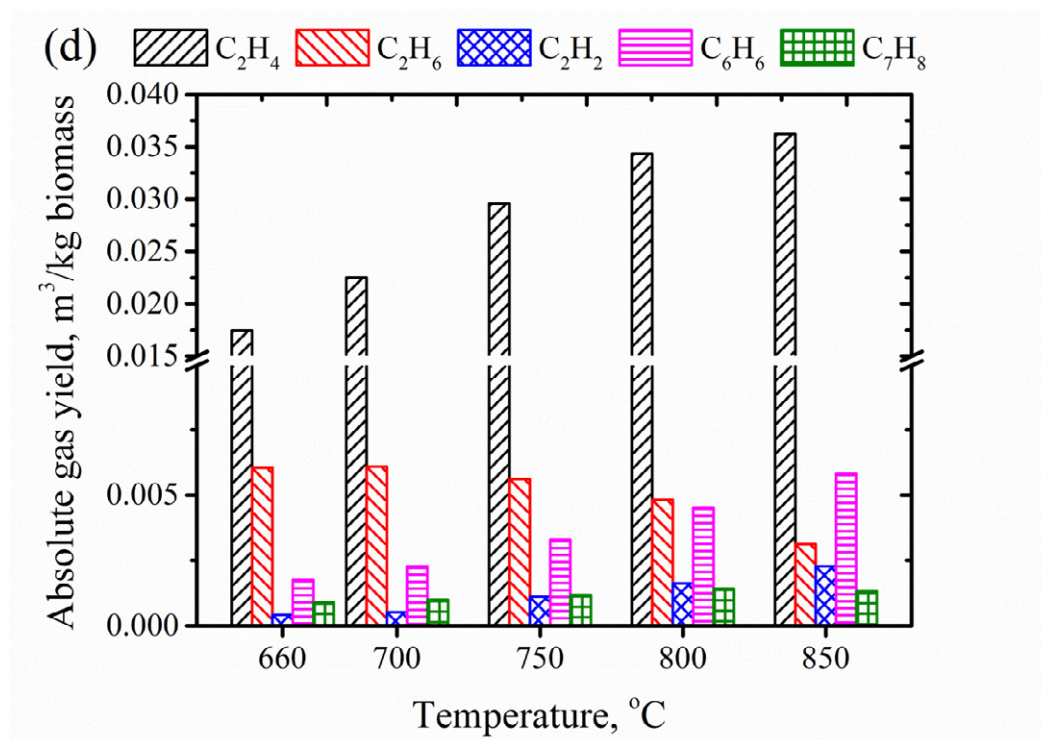


Figure 3 (d)

**Table 1.**

Proximate analysis (as received) (wt.%)	Raw	Torrefied
Moisture	8.76	2.41
Volatile Matter	77.78	69.49
Ash	2.78	4.21
Fixed Carbon	10.68	23.89
Ultimate analysis (dry and ash free basis) (wt.%)		
N	0.66	0.73
C	47.81	54.76
H	6.16	6.09
S	<0.01	<0.01
O	45.37	38.42
HHV (dry basis) (MJ/kg)	17.13	20.90



**Table 2.**

Test	ER	Temperature, °C	Moisture content, vol. %
1	0.175	800	13.50
2	0.206	800	13.05
3	0.232	800	17.78
4	0.255	800	16.93
5	0.199	660	16.77
6	0.202	700	10.72
7	0.202	750	9.82
8	0.202	850	9.69
9	0.318	800	6.54

**Table 3.**

Test No.	Product gas	Elutriated char	Moisture	Sum	Error
1	97.82	2.42	9.99	110.23	-10.23
2	99.17	2.12	9.62	110.91	-10.91
3	94.51	1.71	12.94	109.15	-9.15
4	98.54	1.37	12.56	112.47	-12.47
5	90.83	0.95	11.25	103.03	-3.03
6	102.20	0.86	7.63	110.70	-10.70
7	98.29	1.60	6.74	106.63	-6.63
8	102.12	2.23	7.22	111.56	-11.56
9	100.63	0.36	4.31	105.29	-5.29

Highlights:

1. Torrefied Miscanthus was gasified in an air blown BFB.
2. The gasification conditions were ER (0.177-0.315) and temperature (660-850 °C)
3. The gasification performance of raw and torrefied Miscanthus were compared.
4. Moisture content of product gas was measured.
5. Mass balance of the gasification tests were conducted and compared.